IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of:

Group Art Unit: 10/599,880

WILLIBALD DAFINGER et al.

Examiner: Yate Kai Rene Cutliff

Serial No.: 10/599,880

Filed: February 19, 2007

For: Method For Recovery Of Ethylene In A Recirculating

Gas Process For The Production Of Vinyl Acetate

Attorney Docket No.: WAS 0813 PUSA

APPEAL BRIEF UNDER 37 C.F.R. § 41.37

Mail Stop Appeal Brief - Patents Commissioner for Patents U.S. Patent & Trademark Office P.O. Box 1450 Alexandria, VA 22313-1450

Sir:

This is an Appeal Brief from the final rejection of claims 6 - 14 and 16 of the Office Action mailed on December 4, 2009 for the above-identified patent application.

I. REAL PARTY IN INTEREST

The real party in interest is Wacker Chemie AG ("Assignee"), a corporation organized and existing under the laws of the Germany, and having a place of business at Hanns-Seidel-Platz 4, Munich, Germany, 81737 as set forth in the assignment recorded in the U.S. Patent and Trademark Office on October 14, 2006 at Reel 018390/Frame 0626.

II. RELATED APPEALS AND INTERFERENCES

There are no appeals, interferences or judicial proceedings known to the Appellant, the Appellant's legal representative, or the Assignee which may be related to, directly affect or be directly affected by or have a bearing on the Board's decision in the pending appeal.

III. STATUS OF CLAIMS

Claims 6 - 14 and 16 are pending in this application. Claims 6 - 14 and 16 have been rejected and are the subject of this appeal. Claims 1 - 5 and 16 have been cancelled.

IV. STATUS OF AMENDMENTS

No amendment after final rejection has been filed.

V. SUMMARY OF CLAIMED SUBJECT MATTER

The invention of claim 6 is directed to a process for recovery of ethylene in a recirculating gas process for preparing vinyl acetate, wherein ethylene, acetic acid, and oxygen are reacted at a pressure of 1 - 50 bar and 50 - 200°C in a heterogeneously catalyzed reaction in a reactor, by separating a product gas stream containing ethylene, vinyl acetate, acetic acid, water carbon dioxide, and non-reactive gases, and recycling only a portion of ethylene back to the reactor, by feeding the product gas stream at system pressure directly to a recycle gas scrubber charged with acetic acid, removing vinyl acetate to form a vinyl acetate-free recycle gas, removing CO₂ from this recycle gas to form an ethylenic gas stream, and feeding a portion of the ethylenic gas stream back to the reactor without separating further non-reactive gases, discharging from 1 to 25% of the ethylenic gas stream to another process which consumes ethylene. Specification, page 4, lines 1 - 20, claim 1 as filed, Figure 1.

VI. GROUNDS OF REJECTION TO BE REVIEWED ON APPEAL

Claims 6 - 14 and 16 stand rejected under 35 U.S.C. § 103(a) over Williams et al. EP 0 985 657 ("Williams") in view of Zeyss WO 01/90042 ("Zeyss") in view of Roscher et al. U.S. 4,818,347 ("Roscher") in view of Calcagno et al. U.S. 3,862,216 ("Calcagno") and further in view of Broz U.S. Patent 3,904,656 ("Broz").

VII. ARGUMENT

1. <u>Background</u>

The present invention is directed to the production of vinyl acetate. Vinyl acetate is a commodity chemical produced in large amounts. Hence, as with all high volume commodity chemicals whose processes of preparation are technologically mature, even very small increases in yield, throughput, reduction of reactant costs, minimization of byproducts, etc., are very important commercially, even though when viewed in isolation the improvement might seem small (i.e. <0.1% improvement).

In vinyl acetate production, ethylene, acetic acid, and oxygen react at elevated temperature in the gas phase in a fixed bed reactor. Ethylene conversion is very incomplete, as is also acetic acid conversion, and thus these components must be recycled to the process if economic operation is to be expected. Byproducts of the reaction include relatively large amounts of carbon dioxide as well as methyl acetate and ethyl acetate. In addition, large amounts of "inerts" (inert gases) accumulate, which must be removed from the system. If not removed, the inerts lower vinyl acetate selectivity. The inerts are supplied in numerous sources, for example in the oxygen feed (argon, nitrogen), with the acetic acid (nitrogen) and with the ethylene (methane, ethane).

Due to the low conversion of ethylene and its relatively high cost, the ethylene must be recycled. However, since the product stream contains numerous other substances besides vinyl acetate product and ethylene, including carbon dioxide and inerts, simple recycle has not been possible.

Appellants have improved upon the manufacture of vinyl acetate from the gas phase reaction ethylene and acetic acid by scrubbing the product gas stream with acetic acid, thus removing vinyl acetate from this product stream in a low cost operation, absorbing CO₂ from the recycle vinyl acetate-free stream, also a low cost operation, and then feeding from 75 - 99% of this recycle stream back to the reactor, without separating other components, commonly called inerts. By avoiding the relatively complex and expensive inerts separation, the process economy is markedly increased without affecting the production rate. The remaining 1 - 25% of the vinyl acetate- and CO₂-free ethylene-containing stream is not flared (burned) as in conventional processes, but is routed to another process which employs ethylene as a feedstock, so that the ethylene content is not wasted. The prior art, whether viewed individually or in concert, does not teach or suggest this process, but rather teaches away.

This application has had a prolonged prosecution, and many references have been cited, many dating back to the 1970's, and even the late 1960's. These references together support the need for improvement in the economics of vinyl acetate production, and despite the numerous references cited (and large numbers of other references, many much older), no one has suggested what Appellants claim.

2. <u>Description of the Cited References</u>

a. Williams

That the vinyl acetate process still required improvement is illustrated by Williams et al. published European application EP 0 985 657 ("Williams") (1999). In Williams, the problems of inerts accumulating in the main reactor are thoroughly discussed on page 4. Williams improves upon the prior art by increasing the mol% of ethylene in the reactor to above 60 mol%. However, Williams indicates that due to the high ethylene concentration, inerts are a much more of a serious problem than in other processes which employ low concentrations of ethylene, such as Russell GB 1 264 377. While inerts were tolerable (with decreased selectivity) in Russell, they are intolerable in the Williams' process.

In Russell, for example, inerts, principally nitrogen, constituted about 75 volume percent (and hence roughly 75 mol% also) of the principle reactor contents. Thus, while recycle of a high inerts content recycle stream back to the reactor in Russell will increase the inerts content, it does so only slowly, since the percent inerts in the reactor is already very high and the recycle stream is similar (75% N_2 main reactor feed, 77% recycle stream).

In *Williams*, the ethylene content is much higher, 60 - 85%, and the reactor also contains oxygen and acetic acid. Therefore, the recycle of inerts will cause a serious dilution of the ethylene content. Thus, *Williams* indicates that numerous methods should be used to minimize the amounts of inerts initially introduced, but still requires venting of purge gas to minimize buildup of inerts and CO₂. See paragraph [0028]. Venting (with use of a flare) is very wasteful.

¹This is <u>very</u> dated technology. No one would remotely consider a process such as this, with its low mol% of ethylene, today.

To maximize use of ethylene, *Williams* removes vinyl acetate and acetic acid from the process gas by condensation, removes CO₂ by potash scrubbing, and then separates ethylene from the remaining gases through use of a gas permeable membrane. All the ethylene is then recycled back to the reactor. Inerts must be carefully removed in order to maintain the high mol% of ethylene in the reactor. This is done using the gas permeable membrane.

Thus, Williams recycles all the ethylene in the product gas stream back to the reactor, and is very careful to attempt to remove all inerts prior to doing so. Yet even with membrane purification of the ethylene, Williams still occasionally requires purging gas from the system. Inerts are removed via the purge stream (venting; flare). This is because, like Appellants, Williams operates at high ethylene concentration, See, Williams, page 4 at [0028] and the extensive list of methods to minimize inerts on page 4, lines 16 - 27. The entire thrust of Williams is to operate at very high ethylene concentration which requires minimizing inerts. Williams therefore does not teach or suggest that the ethylene product stream may be recycled without removing inerts, or that only a portion of this stream is recycled. Williams also does not teach or suggest that the ethylene not recycled be used in another process which consumes ethylene. Williams only teaches ([0028]) venting gas from the reactor to remove carbon dioxide and inerts. Since the reactor contains about 60 - 80 volume percent of ethylene, this venting, even though it may only occur periodically, is very wasteful. If it were obvious to direct a crude ethylene stream to another ethylene consuming process, Williams would certainly have mentioned directing a purge stream from the reactor to such a process. However, he does not.

b. Zeyss

Zeyss is directed to a multistage vinyl acetate ("VAM" - vinyl acetate monomer) preparation process wherein in a first stage, ethane (not ethene, or ethylene) is oxidized with an oxygen-containing gas (air) and optionally also in the presence of steam (all examples) to produce a product stream containing ethylene; acetic acid, CO, CO₂, H₂O and other "inerts".

From this stream, CO is removed, leaving a substantial amount of inerts including CO_2 . Additional air $(O_2$, inert (5); see the drawing figure) is added to this stream along with recycle acetic acid and fed to a VAM reactor. As can be seen from Table 1 (page 12), the feed stream contains significant inerts, principally nitrogen ethane, and CO_2 since only the CO has been removed.

Thus, Zeyss process begins with the oxidation of ethane, which may be "high purity ethane" containing 90% ethane. The ethane is catalytically oxidized in a first reactor by addition of air (containing 80% nitrogen) and steam. As a result of this process, not only does the ethane oxidation reactor contain an extremely high proportion of inerts, but the product stream does as well. The Examples of Zeyss may be consulted to verify this large amount of inerts. In all the examples (Table 1), the amount of steam fed to the reactor is of about the same magnitude or more, in most cases, more, than the ethane feed. Also, the amount of air is regulated such that the nitrogen content by volume (and hence by mols also) is 0.8 times the ethane feed. The conversion of ethane is 7.7 to 18.2% (to all products including byproducts - see page 11, next to last paragraph) and the selectivity to ethene is from 0.7 to 59%. Since the ethane feed is always less than 50 mol percent of the total feed and the maximum disclosed selectivity to ethene is about 60%, the product gas will contain but little ethene on a mol basis.

For example, at the highest ethane conversion, 18.2% (Example 11), the feed gas contained 17 volume units (ml/s) of which 6.0 is ethane, a volume (mol) percentage of 35.3% ethane, 6.0 volume parts. At a conversion of 18.2%, this conversion will have consumed 18.2% of the 6.0 parts ethane, leaving 4.9 parts ethane unreacted. Of the 1.1 parts which are converted, 59.0% is converted to ethene, or 0.649 parts. Thus, the ethene feed to the VAM reactor (step b) from the ethane oxidation reactor is about 3.8 mol%! In addition to this feed stream containing only very low amounts of ethene, air (O₂, inert (5)) is also added, as well as a part of the VAM separation (Step (c)) gas stream (6). This latter gas stream has been further depleted of ethene, by the reaction with acetic acid in step (b), so it contains even less than 3.8 mol % of ethene, even

after separating out CO₂. Before routing this gas stream to the ethane oxidation reactor (the gas stream contains more ethane than ethene) or to the VAM reactor, it is treated to remove inerts (see the drawing figure).

Thus, the entire process of Zeyss is simply <u>loaded</u> with inerts. The inerts are predominantly ethane and nitrogen, both present in very large quantities, as well as CO, CO₂ oxygen, other hydrocarbons, etc. This is in direct contrast to Williams, who requires an ethylene concentration in his VAM reactor greater than 60 mol percent, and takes every step to minimize inerts. By contrast, Zeyss purposefully introduces inerts into the VAM reactor, at a very low ethylene concentration.

The entire thrust of Zeyss is to use ethane rather than ethene as a feedstock for VAM production, necessitating a low conversion rate and thus a low concentration of ethene in the VAM reactor. Zeyss apparently overcomes the deficiencies associated with a low ethene content in the VAM reactor by employing a catalyst which is particularly effective in converting ethene and acetic acid to VAM (selectivities 97-98%). However, the salient features of Zeyss are incompatible with the features of Williams, and thus these references are not combinable.

Moreover, although Zeyss discloses use of a scrubber, Zeyss does not indicate that the scrubber is charged with acetic acid, and there is no reason to conclude that this is so. There are numerous scrubbing liquids which may be used. Moreover, the scrubber used by Zeyss is for the separation of ethylene, ethane, inerts CO₂, and CO (page 9 of Zeyss) as an overhead gas stream which is then treated to remove CO₂ and inerts, and this feed stream is fed in large part, to the ethane oxidation reactor, because it contains little ethene, but large amounts of ethane.

3. Roscher

Roscher is directed to the isolation of vinyl acetate from the effluent stream of a gas phase VAM reactor. Roscher teaches that the gas mixture leaving the reactor is passed into a distillation column and fractionated. The gaseous overhead from the column is cooled to a liquid fraction, and gas remaining after cooling is washed with acetic acid in a washing column, to obtain an acetic acid solution containing vinyl acetate. Roscher will be discussed in greater detail later.

4. Calcagno

Calcagno is a completely different vinyl acetate production process from those of Williams, Zeyss, and Roscher, a liquid phase process, which has not been used commercially for years. This process is totally incompatible with the gas phase processes of Appellants, Williams, and the other references. The reaction kinetics are different, the catalyst is different. A portion of the reactor byproduct stream is used to prepare acetic acid, and lower the byproducts in the recycle stream to less than 20 volume percent, an extremely high value. Such a process is incompatible with Williams, who desires an extremely low amount of byproducts and inerts. Calcagno, for example, indicates that the recycle gas, (which forms by far the largest portion of the reactor inlet gas stream since only approximately 2 molar percent of ethylene is converted to vinyl acetate on each pass (column 3, lines 2-3)) is limited to 20 volume percent of oxygen and ethylene, the remaining 80%(!) comprising inert gases (column 2, lines 43-49). This teaching is completely incompatible with Williams.

5. Broz

Broz is cited for use of ethylene as a feed stream for a "further process." Broz does indeed disclose ethylene as a feedstock.

3. The Rejection

The number of references applied to the rejection is evidence alone that the claimed invention is patentable. This is more so the case since there is no evidence of motivation to combine the references, and the references are, for the most part, physically uncombinable. The isolated teachings of the references appear to have been selected based on Appellants' teachings, *i.e.* "picked and chosen" to paraphrase *In re Wesslau*, 353 F.2d 238, 240, 147 USPQ 391, 393 (CCPA 1965).

In the Final Office Action of December 4, 2009, the Examiner stated:

[The] Examiner states that Zeyss was not presented as a process to be combined with the teachings of Williams. Examiner specifically stated that it would have been obvious to one of ordinary skill in the art to, at the time of the present invention was made, to have modified the teaching of Williams by subjecting the product stream gas from the reaction of ethylene, acetic acid and oxygen, to a gas scrubber prior to the removal of the condensable products as suggested by Zeyss. The Examiner directed the Applicant to the fact that the Examiner was focusing on step (b) of Zeyss because it related to the process for producing vinyl acetate, where the reaction products were ethylene, acetic acid and oxygen.

This underscores the problem with the rejection. The rejection <u>is</u> based on a combination of references. In such a combination of references, it is impermissible to pick and choose only so much of a reference as the Office desires to use in formulating a rejection while ignoring the salient teachings of the reference. *Wesslau*, *id*. The fact that the Office has focused on step b) of *Zeyss* is characteristic of an approach which *Wesslau* indicates is improper.

There is no motivation to substitute the optional scrubber of Zeyss, located between the VAM reactor 2 and VAM separator 3 (this optional scrubber referred to by the

Office is not shown in the Figure, but is described on page 8, second full paragraph) for the condenser of *Williams*.

The process of Zeyss, is simply loaded with inerts (as far as the vinyl acetate portion of the process is concerned; (step b) of Zeyss) and operates with a very low concentration of ethylene, since the ethylene is derived as a byproduct from the ethane oxidation (step a)). Williams, on the other hand, operates at a very high ethene concentration and minimizes inerts by first minimizing their input to the reactor as far as is possible (see the preventative methods in Williams in ¶[0028], and eliminates inerts which accumulate despite these measures by purging of the reactor.

While Zeyss teaches an optional scrubbing with acetic acid after his VAM reactor, there is no motivation to use this in the place of Williams' condenser. Moreover, the exit gas from the Zeyss scrubber cannot be used in the Williams' process because it consists substantially of inerts, with very little ethene, but a large amount of ethane. Williams could not tolerate this large amount of inerts in a reactor which must contain minimally 60% ethene and preferably more, along with oxygen and acetic acid, all necessary reactants.

The processes of *Williams* and *Zeyss* are totally different and incompatible. *Williams* operates a VAM reactor with high ethylene feed and is careful to remove inerts from the recycle stream back to the VAM reactor, while *Zeyss* uses an inlet stream to his VAM reactor which has a very low percentage of ethylene and is loaded with inerts. The economy of *Zeyss* is premised on using a cheaper feedstock, ethane, and producing acetic acid, another required reactant, by oxidation of ethane, producing ethylene in the process. *Williams* uses ethylene as the feedstock, not ethane, and preformed acetic acid. These processes are totally different. Why

² The gas stream condensed by *Williams* and the gas stream optionally scrubbed by *Zeyss* have entirely different compositions.

combine them? Where is the evidence of motivation to combine?³ Merely because Zeyss discloses a scrubber does not mean that one would be motivated to use it in the process of Williams. Gas scrubbers and many, many different process apparatuses are described in every chemical engineering textbook. Does this mean that it would be obvious to substitute any of these for any of the unit processes of Williams? There must be some motivation to do so. Williams' system is specifically tailored for production of VAM from a high concentration of ethylene, with severe minimization of inerts. He has described a system suitable for doing so. Why change it? The optional scrubber in Zeyss is used in a process which is far removed from that of Williams. Rather than ethylene dominating the gas in the Zeyss VAM reactor, the gas is dominated by ethane, nitrogen, and water, all inerts. Zeyss proposes a scrubber for use in this highly different environment (although only optionally). Why would this disclosure of Zeyss motivate someone skilled in the art to use such a scrubber in the process of Williams? The answer is that there is no motivation to do so.

Moreover, as can be seen from the process schematic of Zeyss, before the gas stream from the VAM reactor is recycled, it is separated of carbon dioxide and inerts. Appellants' claims require separation of carbon dioxide, but not inerts. Even if there were motivation to combine Zeyss with Williams, the combination teaches removal of inerts, which is not within the scope of Appellants' claims. Both Williams and Zeyss teach removal of inerts. This is a salient feature of Williams; his whole process is premised on minimizing inerts in order to maintain a high ethylene concentration.

Thus, even were *Williams* and *Zeyss* combinable, the combination does not teach or suggest the invention. Once again, it is emphasized, per *Wesslau* that one cannot take Appellants' invention and reconstruct it piecemeal by picking and choosing only so much of the

³ As stated by the Federal Circuit in *In re Dembiczak*, 175 F.3d 994 (Fed. Cir. 2002), evidence of motivation to combine must be present, and this evidence must be "clear and particular."

references as to support a rejection. One cannot simply pick and choose the scrubber of *Zeyss* without also considering his invention as a whole, and there must be some cogent reason to substitute an element from one reference for an element from another reference.

Roscher also appears uncombinable, and if anything, teaches <u>away</u> from the claimed invention. Rather than initially condense the reactor offgas as does *Williams*, *Roscher* introduces the offgas to a distillation column 7, from which gaseous products are taken as an overhead through line 8 through heat exchanger 10, thence to condenser 12, and the gas thus cooled is finally washed in a scrubber 26 with acetic acid. The effluent gas, containing ethylene, oxygen, and carbon dioxide and inerts, col. 6, lines 1 - 2; is recycled to the reactor.

Roscher is apparently cited for use of acetic acid in a gas scrubber. However, Roscher positions his gas scrubber in an entirely different location than Appellants, and in a different location from Zeyss as well. In Roscher, the gas scrubber is located after a first distillation column, a heat exchanger, and a condenser, i.e. far down the offgas processing pathway. Roscher requires this location in his invention because his process is designed to remove as much byproduct ethyl acetate from the process as is possible, at an early stage. Roscher requires the offgas to be first routed to the distillation column. Thus, Roscher teaches against positioning a scrubber immediately downstream from the VAM reactor as Appellants have done.

The use of an acetic acid charged scrubber cannot be simply "picked and chosen" from *Roscher* and combined with *Williams* and *Zeyss*. If there were motivation to "modify" the *Williams* process with the process of *Roscher* (there is no motivation to do this [*Dembiczak*], the *Williams* high ethylene content VAM reactor offgas would be fed to a distillation column (*Roscher*) and the overhead gas would be cooled and condensed, then scrubbed with acetic acid and recycled. However, this is not Appellants' process. Appellants forward the offgas

immediately, at system pressure, to an acetic acid scrubber. Neither *Williams, Zeyss, Roscher*, or their combination teach or suggest this process.

Calcagno is cited, apparently, to teach purification of ethylene. However, Calcagno does not do so. Calcagno is directed to a completely different, liquid phase VAM process which is conducted in the presence of halide salts, which cause halogenated byproducts to be formed. The liquid phase process is an old, outdated process no longer used. Moreover, Calcagno does not teach purification of the ethylene-containing offgas. Calcagno teaches that in prior liquid phase processes this has been attempted, but is "tedious and costly." Calcagno teaches instead to use the ethylene-containing gas stream in a separate reactor to produce acetic acid. Ten percent of the recycle is treated in this way, with the remainder recycled to the VAM reactor. In this way, byproducts in the VAM reactor are limited to 20%, a level much higher than can be tolerated by Williams' gas phase process.

Thus, *Calcagno* teaches against purification of an ethylene recycle stream. Moreover, the impurities in the *Calcagno* recycle stream bear no relationship to the impurities in a gas phase recycle stream. The liquid phase reaction recycle stream contains halogenated hydrocarbons, while the gas phase recycle stream does not. One skilled in the art of gas phase VAM production would not look to *Calcagno* for any purpose. To any extent that *Calcagno* is relevant, he <u>teaches away</u> from the claimed invention.

Broz is cited for using ethylene as a feedstock. Many processes other than VAM production use ethylene as a feedstock. However, there is no teaching or suggestion in *Broz* or any of the other references to employ an ethylene-containing recycle stream from VAM production as an ethylene source.

Broz, for example, is directed to producing ethylene oxide, which is used in large quantities for producing polyoxyethylene polyethers, which are useful as surfactants and as raw

ingredients in the polyol side of polyurethane systems. Copolymer polyols derived by ring opening polymerization of both ethylene oxide and propylene oxide are used in exceedingly large amounts annually. The ethylene oxide used in preparing such polyether polyols and also polyether surfactants,, must be exceptionally pure, which is why the *Broz* process employs such an extensive train of fractional distillation columns in this process.

In ethylene oxide ("EO") production by silver catalyzed epoxidation of ethylene with oxygen, the ethylene oxide product is first washed with water to remove impurities. Some of the EO reacts with water in a ring opening condensation to produce ethylene glycol. Some of the ethylene glycol reacts with one, two, or three further molecules of EO to produce the oligoethers diethylene glycol, triethylene glycol, and tetraethylene glycol. It is an object of *Broz* to obtain the byproduct ethylene glycol in very pure form, so that it can be used to produce fibers, *i.e.* polyethylene terephathalate (polyester) fibers.

Due to the stringent requirements for purity of both the EO and the ethylene glycol, ethylene which is quite pure is used. There is no teaching or suggestion in *Broz* that ethylene recycle from another process could be used in the *Broz* process.

* * * *

Based on the foregoing, it is respectfully submitted that the claimed invention is patentable over the references. The claims are directed to a specific process which the prior art, whether alone or in combination, does not teach or suggest. Not only is the use of a scrubber directly following the VAM reactor and the non-removal of inerts from the recycle stream not taught by the references, but the references also do not teach or suggest feeding the offgas to a scrubber at system pressure, nor has the Office pointed to any portion of the reference which

teaches or suggests this.⁴ None of the references teach or suggest removal of only vinyl acetate and carbon dioxide from the recycle stream. All the references which are pertinent to this aspect of the invention also remove inerts, and *Williams* is, in particular, very adamant in teaching that inerts must be minimized. *Appellants* have discovered a very cost effective process for VAM manufacture which allows for use of the full economic valve of supplied ethylene without removing inerts from the ethylene recycle stream, and yet without allowing inerts to accumulate in the reactor. The benefits of inerts separation is achieved without separation of the inerts, and high efficiency through high reactor ethylene content can be realized in a cost effective manner.

The prior art, whether viewed individually or collectively, does not teach or suggest Appellants' process. Rather, the art teaches away from what Appellants have done. Reversal of the rejection of record is respectfully solicited.

⁴ This is also not inherent, but even if it were, it would not apply, since inherency applies only to rejections under 35 U.S.C. § 102, not § 103. See, *e.g. In re Shetty*, 566 F.2d 81, 195 USPQ 753, 757 (CCPA 1977).

The fee of \$540.00 as applicable under the provisions of 37 C.F.R. § 41.20(b)(2) is being charged to Deposit Account No. 02-3978 via electronic authorization submitted concurrently herewith. The Commissioner is hereby authorized to charge any fees or credit any overpayments as a result of the filing of this paper to Deposit Account No. 02-3978.

Respectfully submitted,

WILLIBALD DAFINGER et al.

William G. Conger

Registration No. 31,209

Attorney/Agent for Applicant

Date: May 3, 2010

BROOKS KUSHMAN P.C.

1000 Town Center, 22nd Floor Southfield, MI 48075-1238

Phone: 248-358-4400 Fax: 248-358-3351

Enclosure - Appendices

VIII. CLAIMS APPENDIX

Claims 1 - 5. (Cancelled)

- 6. A process for ethylene recovery in a recirculating gas process for preparing vinyl acetate, comprising:
 - a) reacting ethylene, acetic acid and oxygen in a heterogeneously catalyzed reaction at a pressure of from 1 to 50 bar and a temperature of from 50°C to 200°C in a reactor,
 - b) separating a product gas stream comprising ethylene, vinyl acetate, acetic acid, water, carbon dioxide and non-reactive gases, and
 - c) recycling only a portion of ethylene back into the reactor, wherein
 - d) the product gas stream is fed from the reactor at system pressure directly to a recycle gas scrubber charged with acetic acid, and vinyl acetate is removed to form a vinyl acetate-free recycle gas, and
 - e) the vinyl acetate-free recycle gas is subsequently fed to a CO₂ absorption to remove carbon dioxide to form an ethylenic gas stream, and then
 - f) a portion of the ethylenic gas stream is recycled into the reactor without further non-reactive gases being separated; and 1% to 25% by volume of the ethylenic gas stream is discharged without further non-reactive gases being separated, and is reused in one or more processes which consume ethylene, other than processes for preparing vinyl acetate.

- 7. The process of Claim 6, wherein the discharged proportion of the ethylenic gas stream is fed to an oxidation processes for preparing ethylene oxide, ethylene glycol, acetaldehyde, or acetic acid, or fed to an oxychlorination of ethylene to dichloroethane, or to a direct chlorination of ethylene to dichloroethane.
- 8. The process of Claim 6, wherein the discharged proportion of the ethylenic gas stream is fed to a process for alkylating benzene to ethylbenzene, a process for carbonylation to acrylic acid, to a polyolefin polymerization, to a hydroformylation process to produce propionaldehyde, in the Reppe carbonylation to propionic acid, or to an Alfol process for preparing long-chain primary alcohols.
- 9. The process of Claim 6, wherein step a) is conducted at a pressure in the range of 8 to 12 bar and at a temperature in the range of 130°C to 200°C.
- 10. The process of Claim 6, wherein the vinyl acetate-free product gas contains 60 to 65 weight percent ethylene and 12 to 18 weight percent carbon dioxide, and after CO₂ removal, contains 80 to 83% ethylene and 1 to 4% CO₂.
- 11. The process of Claim 6, wherein 80 to 95% of the ethylenic cycle gas stream is recycled and 5 to 20% is fed to another process consuming ethylene.

- 12. A process for ethylene recovery in a recirculating gas process for preparing vinyl acetate, comprising:
 - a) reacting ethylene, acetic acid and oxygen in a heterogeneously catalyzed reaction in a reactor at a pressure of from 1 to 50 bar and a temperature of from 50°C to 200°C,
 - b) separating a product gas stream comprising ethylene, vinyl acetate, acetic acid, water, carbon dioxide and non-reactive gases, and
 - c) recycling only a portion of ethylene back into the recirculating gas process, wherein
 - d) the product gas stream is fed at system pressure to a recycle gas scrubber charged with acetic acid, and vinyl acetate is removed from the recycle gas, and
 - e) the vinyl acetate-free recycle gas is subsequently fed to a CO₂ absorption to remove carbon dioxide, and then
 - f) a portion of the ethylenic recycle gas stream is recycled into the reaction system, without further non-reactive gases being separated; and 1% to 25% by volume of the ethylenic gas stream is discharged without further non-reactive gases being separated, and is reused in one or more processes which consume ethylene, other than processes for preparing vinyl acetate,

wherein the product gas stream separated from the reactor in step b) comprises:

from 60 to 65% by volume of ethylene,

from 12 to 18% by volume of CO2,

from 5 to 8% by volume of ethane,

from 4 to 9% by volume of oxygen,

from 4 to 6% by volume of nitrogen,

from 1 to 2% by volume of argon, and

from 0.5 to 1% by volume of methane.

- 13. A process for ethylene recovery in a recirculating gas process for preparing vinyl acetate, comprising:
 - a) reacting ethylene, acetic acid and oxygen in a reactor in a heterogeneously catalyzed reaction at a pressure of from 1 to 50 bar and a temperature of from 50°C to 200°C,
 - b) separating a product gas stream comprising ethylene, vinyl acetate, acetic acid, water, carbon dioxide and non-reactive gases, and
 - c) recycling only a portion of ethylene back into the recirculating gas process, wherein
 - d) the product gas stream is fed at system pressure to a recycle gas scrubber charged with acetic acid, and vinyl acetate is removed from the recycle gas, and

- e) the vinyl acetate-free recycle gas is subsequently fed to a CO₂ absorption to remove carbon dioxide, and then
- f) a portion of the ethylenic recycle gas stream is recycled into the reaction system, without further non-reactive gases being separated; and 1% to 25% by volume of the ethylenic gas stream is discharged without further non-reactive gases being separated, and is reused in one or more processes which consume ethylene, other than processes for preparing vinyl acetate,

wherein the ethylene recycle stream, following treatment to remove carbon dioxide in step e), comprises:

from 80 to 83 % by volume of ethylene,

from 1 to 4 % by volume of CO2,

from 2 to 4 % by volume of ethane,

from 3 to 5 % by volume of oxygen,

from 3 to 4 % by volume of nitrogen,

from 0.5 to 1 % by volume of argon, and

from 0.2 to 0.4% by volume of methane.

14. The process of claim 12, wherein the ethylene recycle stream, following treatment to remove carbon dioxide in step e), comprises:

from 80 to 83 % by volume of ethylene,

from 1 to 4 % by volume of CO2,

from 2 to 4 % by volume of ethane,

from 3 to 5 % by volume of oxygen,

from 3 to 4 % by volume of nitrogen,

from 0.5 to 1 % by volume of argon, and

from 0.2 to 0.4% by volume of methane.

15. Cancelled.

16. The process of claim 12, wherein a portion of 1 - 25% by weight of ethylene-containing gas from the recycle gas scrubber in step d) is fed to a plant for the production of acetic acid from ethylene.

IX. EVIDENCE APPENDIX

None.

X. RELATED PROCEEDINGS APPENDIX

None.